



Thermophysical, rheological and electrical properties of mono and hybrid TiB₂/B₄C nanofluids based on a propylene glycol:water mixture

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ABSTRACT

Hybrid nanofluids aim to further improve the characteristics of mono nanofluids. However, experimental studies that jointly explore the physical properties of hybrids and the corresponding mono nanofluids are missing. In this work, mono B₄C and TiB₂ and hybrid TiB₂:B₄C nanoadditives are used for the first time to design nanofluids based on propylene glycol:water 20:80 wt%. The density, isobaric heat capacity, and thermal conductivity of the nanofluids are determined by the oscillating U-tube, differential scanning calorimetry, and transient hot wire methods, respectively. The rheological behaviour is investigated through rotational rheometry. Additionally, surface tension and electrical conductivity are investigated. The B₄C mono nanofluid shows the highest improvements of thermal conductivity (6.0%) and electrical conductivity (70 times higher), but also the highest viscosity increases (51–54%). The hybrid nanofluid presents intermediate values between those of the mono nanofluids for all the properties except dynamic viscosity. Interactions between spherical and sheet-like nanoparticles explain this behaviour.

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1. Introduction

Nanofluids, dispersions of solid nanoparticles in fluids such as water, glycols or oils, were conceived as working fluids with improved thermal properties compared to the base fluid due to the thermal conductivities of solids are orders of magnitude higher than those of liquids. Multiple highly conductive nanostructured materials have been suggested as additives into the cited thermal fluids. Thus, metal oxides (Al₂O₃, CuO, TiO₂), carbon-based structures (multiple wall and single wall carbon nanotubes, fullerenes) and metals (Au, Ag, Cu) lead the list of most used materials [1,2]. Additionally, nano-enhanced fluids have received great consideration in the last two decades due to their potential qualities in other multiple areas such as energy storage, solar absorption, lubrication, medical purposes, and electrical or magnetic applications [3]. This has broadened the range of materials used, in the search to improve other physical properties of interest such as heat capacity, absorption, surface tension, or electrical conductivity, among others. In addition, more recent research pays attention to new type of nanofluids called hybrid nanofluids that consist of dispersions of hybrid structures of various nanoadditives (composites, decorated particles or mixtures) [4,5].

Hybrid nanofluids aim to further enhance the characteristics of mono nanofluids, based on a potential series of favourable synergistic mechanisms achieved between nanoparticles [6,7]. However, detailed experimental studies that jointly explore different physical properties of hybrid nanofluids are missing. The literature often establishes a good performance for a hybrid nanofluid without analysing all the corresponding mono nanofluids (in some cases none of the mono options is included [8], in others only one of them is considered [9], sometimes comparisons are made with literature values [10]). Comprehensive studies comparing the properties of hybrid nanofluids with those of all the corresponding mono nanofluids in identical conditions (purity of nanopowders and base fluid, nanoadditive concentration, nanofluid preparation, measurement methodology) are required to establish whether the hybrids show values proportional to the amount of each single nanoparticle or if the combination of nanoadditives really causes the appearance of synergistic effects.

Water and glycols are among the most widely used base fluids in nanofluid design [2,11]. Water is one of the best candidates as a heat transfer fluid, since it has the most suitable transport properties and a high heat capacity, but its high melting point at 273 K makes its application very limited. The addition of a glycol to water causes a decrease in the melting point in exchange for worse thermal properties. Propylene glycol has similar thermal properties to ethylene glycol (the most common antifreeze liquid), but its much lower toxicity makes it better suited

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for indirect contact with natural resources, drinking water, or food processing [12]. A binary mixture of propylene glycol and water at 20:80 mass ratio offers protection against freezing up to 265.98 K [13], making it suitable as a heat transfer fluid in secondary systems for geothermal, aerothermal or solar energy heating or cooling applications. However, there is scant literature investigating the properties of nanofluids based on this binary mixture [14–16]. Sundar et al. [14] investigated the thermal conductivity and dynamic viscosity of Fe_3O_4 dispersions based on this mixture and reported increases of up to 20.5% and 123%, respectively, for a 0.5 vol% nanoparticle concentration. Sundar et al. [15] also determined the thermal conductivity and dynamic viscosity of nanodiamond nanofluids based on the same base fluid and described increases of 18.8% and 155%, respectively, for a 1 vol% nanoparticle concentration. Leena et al. [16] reported up to 14.5% thermal conductivity enhancement and 70% dynamic viscosity increases for a 0.2 wt% concentration of TiO_2 in propylene glycol:water 20:80 vol%.

Titanium diboride (TiB_2) is a ceramic material with great hardness, resistance to erosion, and stability to oxidation [17] that presents a high thermal conductivity, similar in magnitude to that of some highly conductive metals like Cu or Al [18]. Boron carbide (B_4C) is one of the hardest ceramic materials, very close to diamond, which also has low density, high impact resistance, excellent chemical resistance, and outstanding thermo-electrical characteristics [19,20]. Both ceramics present good thermal stability at elevated temperatures and a high melting point. Song et al. [21] simulated the flow field in U-shaped tube for boron carbide water-based nanofluids and obtained a good transmission fluid performance for the 0.8 wt% nanoparticle concentration. However, despite the interesting properties of TiB_2 and B_4C , there is a lack of experimental analyses on the physical properties of nanofluids constituted by nanoparticles from both materials, as well as on the corresponding hybrid nanofluids.

There is scant literature jointly analysing physical properties of hybrid nanofluids and all corresponding mono nanofluids in identical conditions. Propylene glycol: water 20:80 wt% as base fluid and titanium diboride and boron carbide as nanoadditives has scarcely been used for the design of nanofluids, despite its interesting properties. Therefore, in this work B_4C and TiB_2 mono nanoadditives and $\text{TiB}_2\text{:B}_4\text{C}$ hybrid nanoadditive are used to design new nanofluids based on propylene glycol:water 20:80 wt%. The thermophysical, rheological and electrical profile of all new dispersions is experimentally described for the first time in the literature and make it possible to observe the strengths and weaknesses of each mono nanofluid and the potential appearance of synergistic effects affecting some physical property for the hybrid nanofluid. Thus, the density, isobaric heat capacity, thermal conductivity, rheological behaviour, surface tension, and electrical conductivity of the nanofluids are investigated in the temperature range of 288.15–318.15 K. The morphology and size, chemical composition, and thermal conductivity of the nanopowders are also examined at room temperature.

2. Materials and methods

2.1. Design

Propylene glycol:water in a mixing ratio of 20:80 wt%, PG:W, was used as base fluid. Propylene glycol was provided by Merck (Darmstadt,

Germany) with a purity of 99.5% and water was produced by a Milli-Q 185 Plus system (Millipore Ltd., United Kingdom). Boron carbide, B_4C , titanium diboride, TiB_2 , and hybrid titanium diboride:boron carbide in a mixing ratio of 20:80 wt%, $\text{TiB}_2\text{:B}_4\text{C}$, were used as nanoadditives. The three nanopowders were commercial samples provided by PlasmaChem (Berlin, Germany) with purities greater than 95% as indicated in Table 1 together with other characteristics. The nanoadditive concentration at 2 wt% was selected to design three nanofluids: 2 wt% $\text{B}_4\text{C/PG:W}$, 2 wt% $\text{TiB}_2\text{/PG:W}$ and 2 wt% $\text{TiB}_2\text{:B}_4\text{C/PG:W}$. This mass fraction of B_4C , TiB_2 and $\text{TiB}_2\text{:B}_4\text{C}$ correspond to 0.00813, 0.00455 and 0.00703 volume fractions, considering density values of the nanopowders of $2520 \text{ kg}\cdot\text{m}^{-3}$ [22,23], $4520 \text{ kg}\cdot\text{m}^{-3}$ [24,25] and $2920 \text{ kg}\cdot\text{m}^{-3}$, respectively.

The nanofluids were prepared following a two-step method. The amounts of each base fluid component and each nanoadditive were weighed on a CPA225 balance from Sartorius AG (Goettingen, Germany) with an uncertainty of 0.01 mg. Subsequently, the dispersions were sonicated for 20 min using a Sonopuls HD 2200 ultrasonic homogenizer (Bandelin Electronic, Germany) coupled with a 3 mm diameter titanium tip operating at a frequency of 20 kHz and a power of 200 W. A bath of ice around the sample was used to prevent overheating.

2.2. Experimental

The morphology and size of the dry nanopowders were analysed by scanning electron microscopy (SEM) using a JEOL JSM-6700F field emission scanning electron microscope (JEOL, Tokyo, Japan). The chemical composition was determined by Energy-dispersive X-ray spectroscopy microanalyses (EDS) using an Inca Energy 300 spectrometer (Oxford Instruments, Oxford, UK) coupled to the aforementioned SEM. One drop of each nanopowder dispersion in analytical grade methanol was dried at room temperature on the silica support prior to SEM and EDS measurements.

The thermal conductivity (k) of the nanopowders was determined at room temperature and atmospheric pressure using a DTC-25 thermal conductivity meter (TA instruments, UK). The operation of this device is based on the guarded heat flow meter method, in accordance with the ASTM E1530 standard [26]. The sample discs are prepared by compacting the nanopowder with a manual system press. Then, each sample disk is placed between the heat source located on the top plate and the heatsink located below the bottom plate, and a steady heat flow is established between them. The expanded uncertainty of the assessed thermal conductivity values is declared as 6% [27].

The density (ρ) of the nanofluids and the base fluid was determined in a temperature range of 288.15–318.15 K using a DMA 4100 M density meter (Anton Paar, Austria). The operation of this device is based on the oscillating U-tube method. The sample temperature was stabilized with an accuracy of 0.02 K. The expanded uncertainty of the measured ρ values is declared as 0.1% [28].

The isobaric heat capacity (c_p) of the nanofluids and the base fluid was measured in a temperature range of 288.15–318.15 K using a Q2000 heat flow differential scanning calorimeter (TA Instruments, USA) coupled with a RSC90 cooling system. Measurements are based on the quasi-isothermal temperature modulated differential scanning calorimetry method and were conducted in a nitrogen atmosphere

Table 1

Main characteristics of the commercial B_4C , TiB_2 and $\text{TiB}_2\text{:B}_4\text{C}$ nanopowders according to the manufacturer.

Nanopowder	Provider	Nomenclature	Average particle size	Purity	Controlled admixtures
Boron carbide	PlasmaChem	B_4C	60 nm	>99%	$\text{Fe} < 0.0001\%$, $\text{Ni} < 0.0002\%$, $\text{Al} < 0.0002\%$
Titanium diboride	PlasmaChem	TiB_2	<1 μm	>98.5%	$\text{B}_2\text{O}_3 < 0.1\%$, $\text{C} < 0.3\%$, $\text{Fe} < 0.1\%$, $\text{Si} < 0\%$
Titanium diboride - Boron carbide (20:80)	PlasmaChem	$\text{TiB}_2\text{:B}_4\text{C}$	Composition: $\text{TiB}_2 > 19.7\%$ $\text{B}_4\text{C} > 77\%$	–	$\text{B}_2\text{O}_3 < 0.18\%$, $\text{C} < 2.46\%$, $\text{Fe} < 0.02\%$, $\text{Si} < 0.0\%$

using Tzero pans and Tzero hermetic lids of aluminium (TA Instruments). The temperature was sinusoidally modulated with an amplitude of 0.5 K and a period of 80 s around the selected value. The expanded uncertainty of measured c_p values is declared as 3% [29].

The thermal conductivity (k) of the nanofluids and the base fluid was determined in a temperature range of 288.15–318.15 K using a THW-L2 thermal conductivity meter (Thermtest, Canada) coupled to a Peltier IC20XR dry bath (EchoTherm). Measurements are based on the transient short hot wire (SHW) technique, in accordance with the ASTM D7896–14 standard [30]. A 60 mm long 0.1 mm diameter aluminium wire is inserted vertically into the container with 18 ml of sample inside for measurements. The whole set covered with an insulating box is placed on the dry bath. An initial power test adjusts the heat power required to produce the local temperature rise necessary for the measurement in a short period of time, avoiding natural convection [28,31]. The expanded uncertainty of the measured k values is declared as 5% [28].

The rheological characterization of the nanofluids and the base fluid was determined in a temperature range of 288.15–318.15 K using a HAAKE MARS 2 rheometer (Thermo Electron Corporation, Germany) coupled to a Peltier system associated to a Phoenix 2 thermostat (Thermo Electron Corporation), ensuring the establishment of the target temperature with 0.1 K uncertainty. A DC60/1 double cone geometry (60 mm diameter, 1° cone angle) was used to perform rotational measurements at shear rate range from 10 s^{-1} to 1000 s^{-1} , 10 points per decade. The dynamic viscosity (η) values reported here were obtained as the mean value in the shear rate range $100\text{--}1000 \text{ s}^{-1}$, where all samples behave in a Newtonian manner. The expanded uncertainty of the presented η values is declared as 5% [32,33].

The surface tension (γ) between air and nanofluid/base fluid was determined in a temperature range of 288.15–318.15 K using a tensiometer PI-MT1A (Polon-Izot, Warsaw, Poland). The measurements of this device are based on the Du Noüy ring method. The sample temperature was stabilized with an accuracy of 0.02 K. More information on the measurement procedure can be found in Ref. [34]. The expanded uncertainty of the measured γ values is declared as 1% [34].

The electrical conductivity (σ) of the nanofluids and the base fluid was determined in a temperature range of 288.15–318.15 K using a Multiline 3410 conductivity meter (WTW GmbH, Germany) with TetraCone 925 conductivity probe. The temperature was stabilized for at least 10 min with a self-made heating/cooling chamber with 0.2 K accuracy. The expanded uncertainty of the measured σ values is declared as 0.5% [35].

3. Results

3.1. Nanopowder characterization

Fig. 1 contains SEM images of the used nanoadditives, evidencing the differences on shape and size between them. Fig. 1a shows quasi-spherical B_4C nanoparticles with radii close to 60 nm, while Fig. 1b shows irregularly shaped TiB_2 sheets with lengths/widths generally less than $1 \mu\text{m}$. Fig. 1c shows B_4C nanoparticles deposited on the surface of TiB_2 sheets, with both nanoparticles with the same sizes and shapes previously described. The obtained sizes by means these figures agree with those declared by the manufacturer (Table 1).

The EDS spectrums of the nanopowders used are presented in Fig. 2. The results obtained for B_4C (Fig. 2a) confirm the majority presence of carbon (C) and boron (B), as expected, and the residual appearance of oxygen (O). The results for TiB_2 (Fig. 2b) show the majority presence of titanium (Ti) and boron (B), with a negligible appearance of aluminium (Al). The results for $\text{TiB}_2\text{:B}_4\text{C}$ (Fig. 2c) show the majority presence of boron (B), carbon (C) and titanium (Ti), with small percentages of oxygen (O) and iron (Fe), which agrees with the manufacturer's information (Table 1). The appearance of silica (Si) in all spectra is explained by the physical support on which the analyses were made.

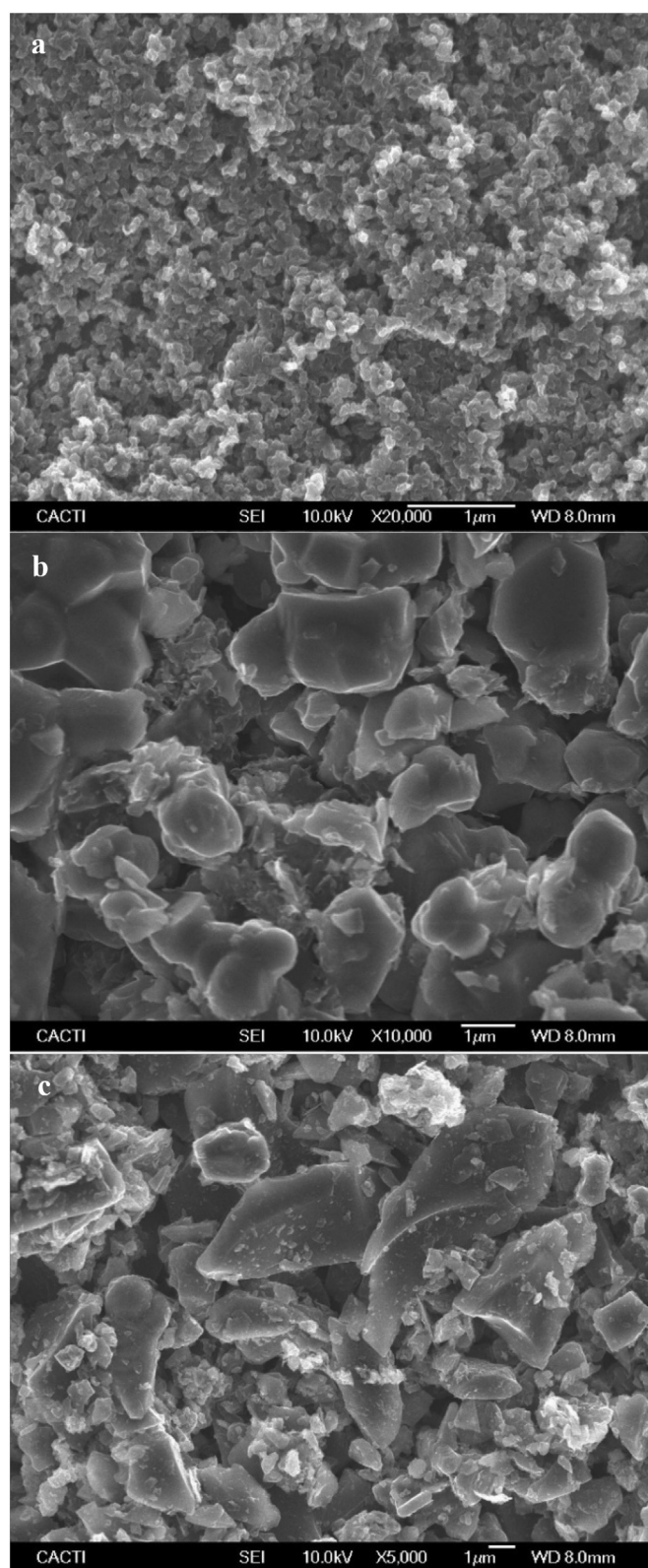


Fig. 1. SEM images of B_4C (a), TiB_2 (b) and $\text{TiB}_2\text{:B}_4\text{C}$ (c) nanopowders.

3.2. Density

Table 2 and Fig. 3 show the density values obtained for the base fluid and the nanofluids as a function of temperature. The experimental values obtained for PG:W reach a maximum deviation with respect to

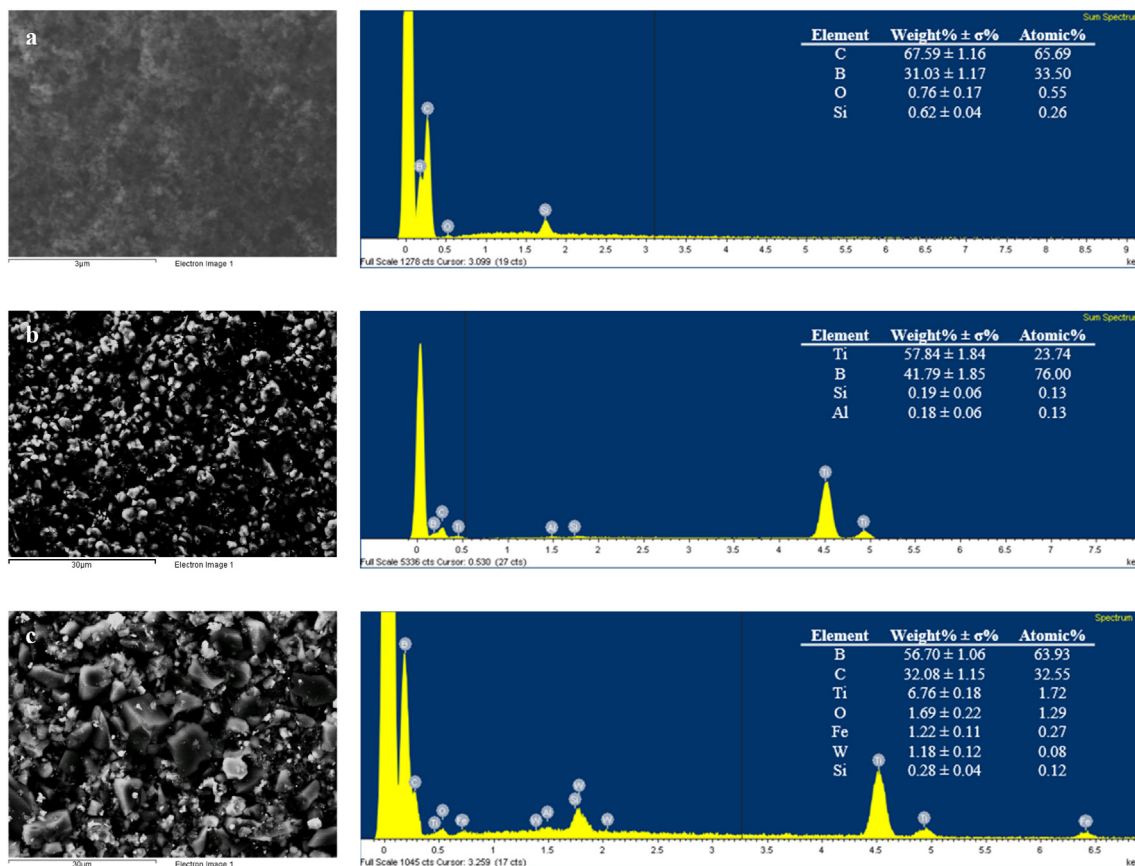


Fig. 2. EDS microanalyses of B₄C (a), TiB₂ (b) and TiB₂:B₄C (c) nanopowders.

the literature [13] of 0.02%. Identical density drops of 1.2% are detected for all samples when the temperature increases from 288.15 to 318.15 K, which shows that the addition of nanoadditives does not alter this behaviour.

However, the dispersion of the different nanoadditives produces dissimilar increases in density with respect to the base fluid. Rises with respect to PG:W of 0.95%, 1.5% and 1.1% are observed for the B₄C, TiB₂ and TiB₂:B₄C nanofluids, respectively. The dispersion of the mono nanoadditive TiB₂ entails higher density increases than the dispersion of the mono nanoadditive B₄C, in accordance with the density values of the corresponding bulk materials, 4520 kg·m⁻³ [24,25] and 2520 kg·m⁻³ [22,23], respectively. The hybrid nanoadditive TiB₂:B₄C entails a density increase closer to that of B₄C, in agreement with the mixing ratio between materials, 20:80 wt%. It can also be noticed that the increases in density due to the dispersion of nanoadditives are temperature-independent in the range analysed.

Pak and Cho [36] used the classical mixing theory for ideal gas mixtures to estimate the density of nanofluids, considering them simple solid-liquid mixtures:

Table 2

Experimental densities^a, ρ , for the PG:W base fluid and the B₄C, TiB₂ and TiB₂:B₄C nanofluids at different temperatures^b, T .

T [K]	ρ [kg·m ⁻³]			
	0 wt%	2 wt% B ₄ C	2 wt% TiB ₂	2 wt% TiB ₂ :B ₄ C
288.15	1016.2	1025.8	1031.9	1027.1
298.15	1012.7	1022.3	1028.4	1023.6
308.15	1008.5	1018.1	1024.2	1019.4
318.15	1003.7	1013.2	1019.2	1014.5

^a Expanded uncertainty ($k = 2$) of density: 0.1%.

^b Expanded uncertainty ($k = 2$) of temperature: 0.1 K.

$$\rho_{nf} = \phi_v \cdot \rho_{nf} + (1 - \phi_v) \cdot \rho_{bf} \quad (1)$$

where ϕ_v means nanoadditive volume fraction, and the subscripts nf, np and bf mean nanofluid, nanopowder and base fluid, respectively. The ρ_{np} value generally used for calculations is that of the corresponding bulk solid.

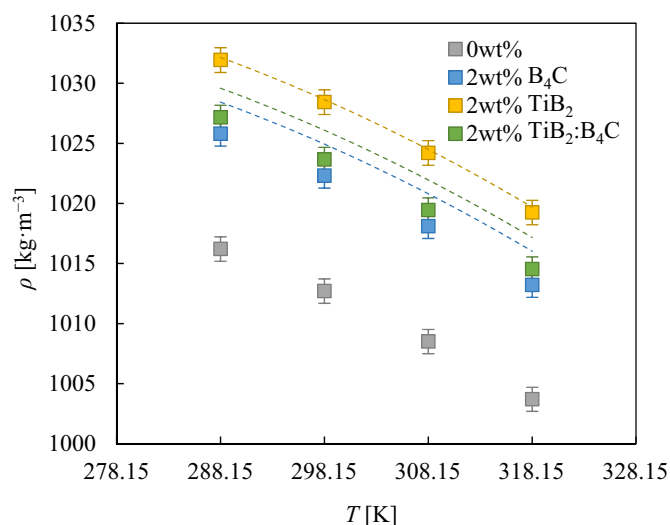


Fig. 3. Dependence of density, ρ , on temperature, T , for the PG:W base fluid and the 2 wt% B₄C, TiB₂ and TiB₂:B₄C nanofluids. Pak and Cho model, Eq. (1) (—).

A good agreement was obtained between the experimental and predicted data using ρ_{np} values of $4520 \text{ kg} \cdot \text{m}^{-3}$ [24,25], $2520 \text{ kg} \cdot \text{m}^{-3}$ [22,23] and $2920 \text{ kg} \cdot \text{m}^{-3}$ for TiB_2 , B_4C and $\text{TiB}_2\text{:B}_4\text{C}$, respectively. Average deviations of -0.26% , -0.03% and -0.05% were obtained, respectively, as can be seen in Fig. 3.

3.3. Isobaric heat capacity

Table 3 and Fig. 4 show the isobaric heat capacities for the base fluid and the nanofluids as a function of temperature. The experimental values obtained for PG:W reach a maximum deviation with respect to the literature [13] of 1.3%. Similar heat capacity increases of 1.6% are noticed for all samples when the temperature increases from 288.15 to 318.15 K.

The dispersion of the nanoadditives within the base fluid leads to c_p average decreases of 1.51%, 1.70% and 1.54% for B_4C , TiB_2 and $\text{TiB}_2\text{:B}_4\text{C}$, respectively. The dispersion of the mono nanoadditive TiB_2 produces higher c_p decreases than the dispersion of the mono nanoadditive B_4C , in agreement with the fact that the bulk solid c_p is lower for TiB_2 , $617 \text{ J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ [37,38], than for B_4C , $950 \text{ J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ [39,40]. The dispersion of the hybrid nanoadditive $\text{TiB}_2\text{:B}_4\text{C}$ entails an intermediate c_p decrease with a value closer to that of B_4C , in agreement with the mixing ratio between materials, 20:80 wt%. Similar c_p decreases are observed due to the dispersion of nanoadditives for all the temperatures analysed.

Xuan and Roetzel [41,42] applied the theory of mixtures to estimate the isobaric heat capacity of nanofluids on the basis of the mass fraction of nanoparticles in the dispersion:

$$c_{p \text{ nf}} = \phi_m \cdot c_{p \text{ np}} + (1 - \phi_m) \cdot c_{p \text{ bf}} \quad (2)$$

where ϕ_m means nanoadditive mass fraction. The $c_{p \text{ np}}$ value usually used for calculations is that of the corresponding bulk solid.

Good agreement was obtained between the experimental and predicted data using $c_{p \text{ np}}$ values of $617 \text{ J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ [37,38], $950 \text{ J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ [39,40] and $883 \text{ J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ for TiB_2 , B_4C and $\text{TiB}_2\text{:B}_4\text{C}$, respectively, with absolute deviations lower than 0.5%, as can be observed in Fig. 4.

3.4. Thermal conductivity

Table 4 and Fig. 5 gather the thermal conductivities for the base fluid and the nanofluids as a function of temperature. The experimental values obtained for PG:W reach a maximum deviation with respect to the literature [13] of 0.85%. Thermal conductivity increases between 5.6 and 6.0% are observed when the temperature increases from 288.15 to 318.15 K.

As regards the comparison between nanofluids and base fluid, the mono nanoadditive B_4C produces the highest thermal conductivity enhancements, $\sim 6.0\%$, followed by the hybrid nanoadditive $\text{TiB}_2\text{:B}_4\text{C}$, $\sim 4.5\%$. In contrast, the mono nanoadditive TiB_2 produces the lowest increases, $\sim 1.2\%$. As observed, the $\text{TiB}_2\text{:B}_4\text{C}$ hybrid nanoadditive involves an intermediate k increase between those of the corresponding mono nanoadditives. It should be noted the k

Table 3

Experimental isobaric heat capacities^a, c_p , for the PG:W base fluid and the B_4C , TiB_2 and $\text{TiB}_2\text{:B}_4\text{C}$ nanofluids at different temperatures^b, T .

T [K]	$c_p [\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}]$			
	0 wt%	2 wt% B_4C	2 wt% TiB_2	2 wt% $\text{TiB}_2\text{:B}_4\text{C}$
288.15	4018	3956	3949	3955
298.15	4035	3974	3966	3973
308.15	4056	3995	3988	3994
318.15	4081	4020	4013	4019

^a Expanded uncertainty ($k = 2$) of isobaric heat capacity: 3%.

^b Expanded uncertainty ($k = 2$) of temperature: 0.1 K.

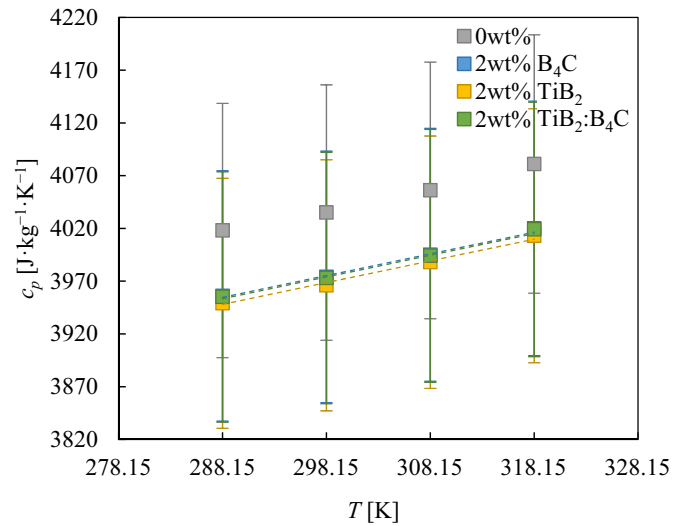


Fig. 4. Dependence of isobaric heat capacity, c_p , on temperature, T , for the PG:W base fluid and the B_4C , TiB_2 and $\text{TiB}_2\text{:B}_4\text{C}$ nanofluids. Xuan and Roetzel model, Eq. (2) (—).

Table 4

Experimental thermal conductivities^a, k , for the PG:W base fluid and the B_4C , TiB_2 and $\text{TiB}_2\text{:B}_4\text{C}$ nanofluids at different temperatures^b, T .

T [K]	$k [\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}]$			
	0 wt%	2 wt% B_4C	2 wt% TiB_2	2 wt% $\text{TiB}_2\text{:B}_4\text{C}$
288.15	0.491	0.520	0.497	0.514
298.15	0.502	0.532	0.507	0.524
308.15	0.512	0.543	0.519	0.536
318.15	0.520	0.551	0.526	0.543

^a Expanded uncertainty ($k = 2$) of thermal conductivity: 3%.

^b Expanded uncertainty ($k = 2$) of temperature: 0.2 K.

enhancements due to the dispersion of nanoadditives are practically temperature-independent in the range analysed. It can be also noticed that the order between the thermal conductivity increases is the same as that established between the thermal conductivity values of the nanopowders obtained by the guarded heat flow method, 5.0, 4.4 and $1.4 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ for B_4C , $\text{TiB}_2\text{:B}_4\text{C}$ and TiB_2 , respectively.

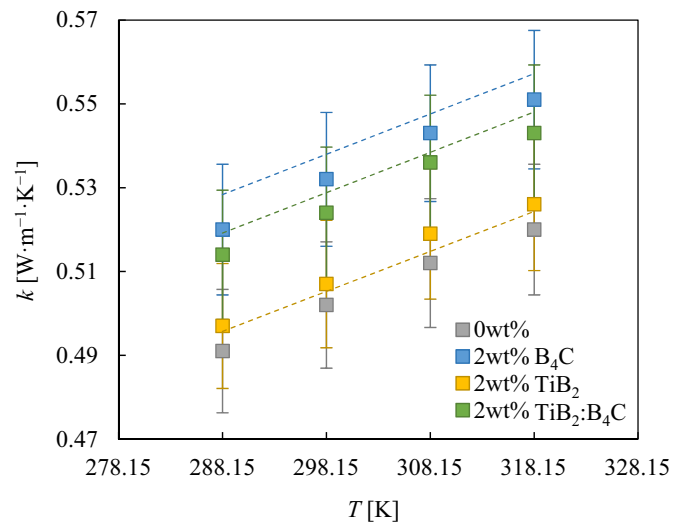


Fig. 5. Dependence of thermal conductivity, k , on temperature, T , for the PG:W base fluid and the B_4C , TiB_2 and $\text{TiB}_2\text{:B}_4\text{C}$ nanofluids. Parallel model, Eq. (3) (—).

The parallel model or upper Wiener bound [43] for determining the thermal conductivity of mixtures has been previously adapted by our group to the field of nanofluids [12,27] according to the following equation:

$$k_{nf} = \phi_v \cdot k_{np}(1 - \phi_v) \cdot k_{bf} \quad (3)$$

In this case, the k_{np} value used in calculations is the experimental thermal conductivity of the nanopowder here obtained.

Absolute deviations lower than 1.5% were obtained between the experimental and predicted data using the aforementioned nanopowder thermal conductivities, as can be observed in Fig. 5.

The thermal conductivity results here reported can be compared with those previously published by Sundar et al. [14,15] for mono nanofluids based on the same base fluid, propylene glycol:water 20:80 wt%. They obtained increases of 10.4% and 9.9% for 0.5 vol% concentration of Fe_3O_4 [14] and 1 vol% concentration of nanodiamonds [15] at 293.15 K, respectively. At that temperature, these literature results are around 4% and 9% higher than those of the 2 wt% B_4C and TiB_2 mono nanofluids, respectively, and around 5.5% higher than those of the 2 wt% $\text{TiB}_2\text{:B}_4\text{C}$ hybrid nanofluid. On the other hand, the non-dependence on temperature of the thermal conductivity improvement reported here was not observed in the aforementioned previous works, since in both cases thermal conductivity shows huge enhancements with the temperature increase.

3.5. Rheological behaviour

The flow curves obtained for the base fluid and the nanofluids at different temperatures are presented in Fig. 6. A constant viscosity value for all the analysed shear rates clearly evidences the Newtonian behaviour of the base fluid. Contrastingly, the nanofluids show a non-Newtonian pseudoplastic or shear thinning behaviour. This behaviour is evidenced at the lower shear rates analysed, while a Newtonian plateau appears at the higher shear rates values. The shear thinning behaviour of nanoadditive dispersions has been attributed in the literature [32,44] to modifications in the arrangement of the particles within the base fluid. Nanoadditive agglomerates could break once oriented in the flow direction of the shear, reducing the amount of solvent bound among nanoparticles. Thus, the interaction forces in the dispersion would weaken, reducing its resistance to flow, and accordingly its apparent viscosity at higher shear rates [32,44]. It can be also observed that titanium diboride and boron carbide nanofluids exhibit stronger viscoelastic structure at lower temperatures, in accordance with previous results on carbon-based nanofluids [33].

Table 5 and Fig. 7 summarize the dynamic viscosities for the base fluid and the nanofluids obtained as the average value in the shear rate range from 100 s^{-1} to 1000 s^{-1} of the flow curves, where all samples reach the second Newtonian plateau. The obtained values for PG:W reach a maximum deviation with respect to the literature [13] of 4.1%. Similar dynamic viscosity decreases with the increasing temperature were detected for all samples, 54–56%, indicating that the addition of nanoadditives does not significantly alter this temperature-dependent behaviour.

Vogel–Fulcher–Tammann (VFT) equation [45–47] was used to correlate the dependence of the dynamic viscosity on temperature:

$$\eta = \eta_0 \cdot e^{\frac{A \cdot T_0}{T - T_0}} \quad (4)$$

where η_0 , A and T_0 are the fitting parameters. Absolute average deviations lower than 0.6% were obtained and the adjustment parameters are gathered in Table 5.

The different nanoadditives produces dissimilar dynamic viscosity rises with respect to the base fluid. Increases with respect to base fluid up to 51–54%, 17–24% and 10–14% are observed for the B_4C , TiB_2 and $\text{TiB}_2\text{:B}_4\text{C}$ nanofluids, respectively. The mono nanoadditive B_4C implies the highest dynamic viscosity increases and, in contrast to the thermophysical properties presented above, the hybrid nanoadditive $\text{TiB}_2\text{:B}_4\text{C}$ does not entail an intermediate variation between the two mono nanoadditives, but produces the least dynamic viscosity increase. This fact implies the appearance of a beneficial effect in terms of flow behaviour between the two types of TiB_2 and B_4C nanoparticles. Possible interactions between spherical and sheet-like nanoparticles could explain this lower resistance to flow.

There is scant literature comparing the viscosity of nanofluids containing hybrid nanoadditives with those containing the corresponding mono nanoadditives, and more studies are required on this subject as recognized by a recent review paper [48]. Gangadevi and Vinayagam [49] compared the dynamic viscosity of $\text{Al}_2\text{O}_3/\text{water}$, CuO/water and $\text{Al}_2\text{O}_3\text{:CuO}/\text{water}$ (50:50 wt%), obtaining the lower viscosity increases for the hybrid nanofluid at 0.1 wt% total nanoadditive concentration. Among the existing studies comparing hybrid nanofluids with different mixing ratios between nanomaterials, several experimental studies [50,51] found that the relationship between viscosity and mixing ratios is observably non-linear, with some intermediate mixing ratio obtaining lower dynamic viscosity increases than the others. The authors explain this fact by the different mechanisms on the shearing flow resistance due to the presence of two types of nanoparticles with different particle sizes and shapes [50,51].

The dynamic viscosity results reported here can be compared with those previously noticed by Sundar et al. [14,15] for mono nanofluids based on the same base fluid, propylene glycol:water 20:80 wt%. They

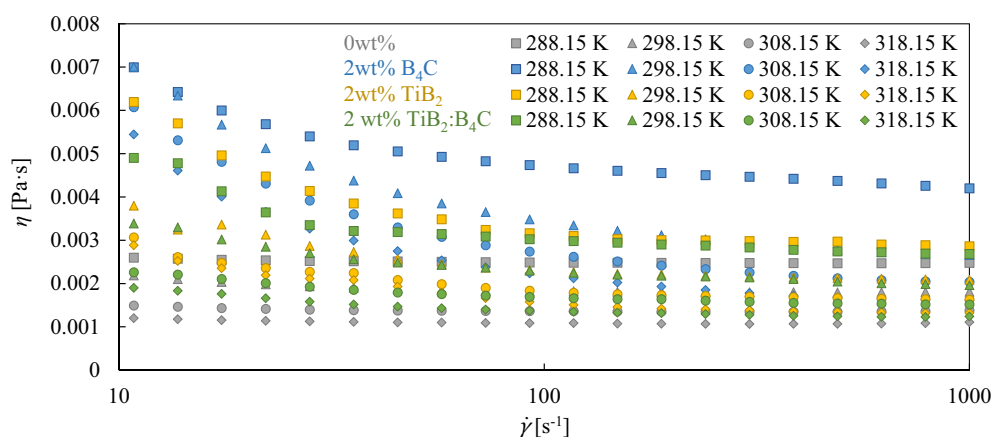


Fig. 6. Dependence of dynamic viscosity, η , on shear rate, $\dot{\gamma}$, for the base fluid and the B_4C , TiB_2 , and $\text{TiB}_2\text{:B}_4\text{C}$ nanofluids.

Table 5

Experimental dynamic viscosities^a, η , for the PG:W base fluid and the B₄C, TiB₂ and TiB₂:B₄C nanofluids at different temperatures^b, T , and fitting parameters, η_0 , A and T_0 , standard deviations, s , and absolute average deviations, AAD, from Vogel–Fulcher–Tammann equation, Eq. (4).

T [K]	η [mPa·s]			
	0 wt%	2 wt% B ₄ C	2 wt% TiB ₂	2 wt% TiB ₂ :B ₄ C
288.15	2.47	3.74	2.92	2.72
298.15	1.80	2.74	2.11	2.02
308.15	1.36	2.09	1.65	1.53
318.15	1.08	1.66	1.34	1.24
η_0 [mPa·s]	0.0386	0.0522	0.2148	0.0421
A	2.984	3.382	0.851	3.313
T_0 [K]	167.8	160.8	217.3	160.6
s [mPa·s]	0.008	0.006	0.008	0.022
AAD	0.23%	0.11%	0.21%	0.60%

^a Expanded uncertainty ($k = 2$) of dynamic viscosity: 3%.

^b Expanded uncertainty ($k = 2$) of temperature: 0.1 K.

obtained increases of around 34% and 35% for a 0.5 vol% concentration of Fe₃O₄ [14] and a 1 vol% concentration of nanodiamonds [15] at 293.15 K, respectively. At that temperature, these results are around 20% lower than those of the 2 wt% B₄C mono nanofluid and around 10% and 20% higher than those of the 2 wt% B₄C mono nanofluid and the 2 wt% TiB₂:B₄C hybrid nanofluid, respectively.

3.6. Surface tension

Table 6 and Fig. 8 show the temperature dependence of the surface tension of three types of nanofluids containing B₄C, TiB₂, TiB₂:B₄C mixture and pure base fluid. It can be observed that the surface tension of the tested nanofluids does not differ much from the binary PG:W base fluid.

On the other hand, as the temperature increases, more and more differences between the tested samples are visible. For sample with B₄C nanoparticles, the decrease in surface tension with temperature occurs to a lesser extent than for the other samples which is most noticeable at 318.15 K. Similar surface tension variations were observed for metal oxide-based and nitride-based nanofluids [34,52,53].

3.7. Electrical conductivity

Table 7 and Fig. 9 present the dependence of electrical conductivity on temperature for the three nanofluids containing B₄C, TiB₂, and their hybrid, and for the PG:W base fluid.

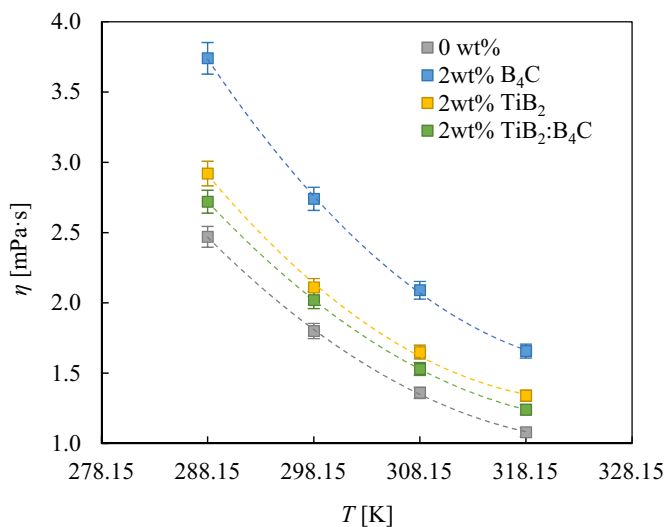


Fig. 7. Dependence of dynamic viscosity, η , on temperature, T , for the PG:W base fluid and the B₄C, TiB₂ and TiB₂:B₄C nanofluids. Vogel–Fulcher–Tammann equation, Eq. (4) (—).

Table 6

Experimental surface tensions^a, γ , for the PG:W base fluid and the B₄C, TiB₂ and TiB₂:B₄C nanofluids at different temperatures^b, T .

T [K]	γ [N·m ⁻¹]			
	0 wt%	2 wt% B ₄ C	2 wt% TiB ₂	2 wt% TiB ₂ :B ₄ C
288.15	0.05738	0.05762	0.05692	0.05744
298.15	0.05579	0.05631	0.05588	0.05589
308.15	0.05408	0.05510	0.05433	0.05429
318.15	0.05234	0.05360	0.05218	0.05244

^a Expanded uncertainty ($k = 2$) of surface tension: 1%.

^b Expanded uncertainty ($k = 2$) of temperature: 0.1 K.

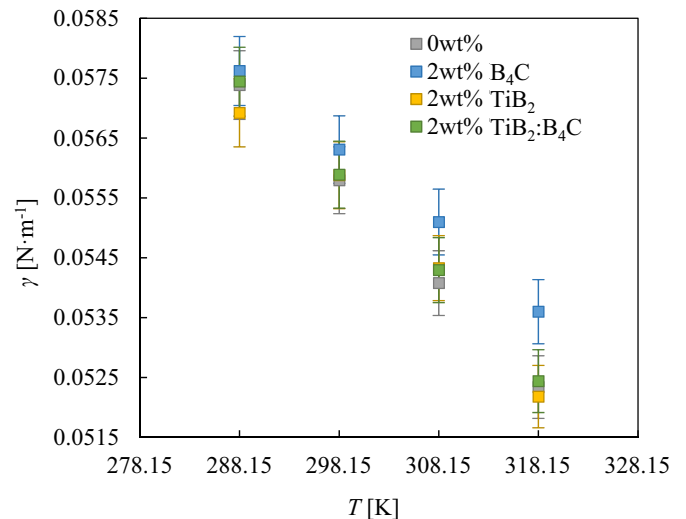


Fig. 8. Dependence of surface tension, γ , on temperature, T , for the PG:W base fluid and the B₄C, TiB₂ and TiB₂:B₄C nanofluids.

The highest enhancement with temperature increase was observed in the case of nanofluid with B₄C nanoparticles, where noted increase from 2.66 $\mu\text{S}\cdot\text{cm}^{-1}$ to 208.2 $\mu\text{S}\cdot\text{cm}^{-1}$ at the tested temperature range. Likewise, this sample has the highest value of electrical conductivity among the tested nanofluids with values up to 70 times higher than the base fluid. On the other hand, the electrical conductivity of TiB₂ nanofluid is much lower vulnerable to the effect of temperature. Observed changes with temperature increase are nearly ten times smaller than that B₄C nanofluid. Also, the nanofluid prepared with the mixture of B₄C and TiB₂ particles show similar behaviour as TiB₂ nanofluid, which suggests a negative effect of bigger particles TiB₂ on the electrical conductivity of B₄C nanofluid. A similar effect was observed by Khalid et al. [54] for hybrid nanofluids containing SiO₂ and Al₂O₃ nanoparticles in ethylene glycol. The effect was explained by the lower electrical conductivity of the nanofluids produced with Al₂O₃ than SiO₂ nanoparticles. In the case of this study, the significant difference in electrical conductivity for B₄C nanofluid and their hybrid

Table 7

Experimental electrical conductivities^a, σ , for the PG:W base fluid and the B₄C, TiB₂ and TiB₂:B₄C nanofluids at different temperatures^b, T .

T [K]	σ [$\mu\text{S}\cdot\text{cm}^{-1}$]			
	0 wt%	2 wt% B ₄ C	2 wt% TiB ₂	2 wt% TiB ₂ :B ₄ C
288.15	2.66	208.2	20.91	22.91
298.15	4.00	273.6	28.80	31.37
308.15	5.59	345.2	37.22	40.82
318.15	7.37	419.9	46.45	50.74

^a Expanded uncertainty ($k = 2$) of electrical conductivity: 1%.

^b Expanded uncertainty ($k = 2$) of temperature: 0.1 K.

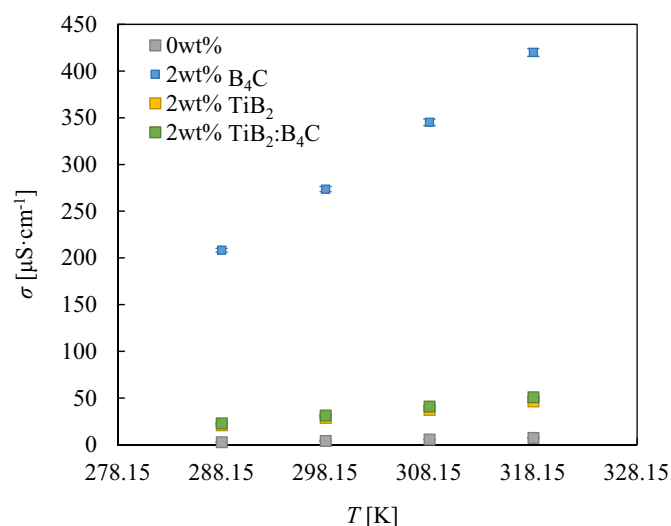


Fig. 9. Dependence of electrical conductivity, σ , on temperature, T , for the PG:W base fluid and the B_4C , TiB_2 and $TiB_2:B_4C$ nanofluids.

with TiB_2 can be caused by the morphology of mixed powders where smaller particles of B_4C are deposited on much larger particles of TiB_2 in effect, the active surface area of particles is significantly lower than that for mono B_4C particles.

4. Conclusions

Hybrid nanofluids aim to further improve the characteristics of mono nanofluids based on potential synergistic mechanisms between nanoparticles. Comprehensive studies comparing each physical property of hybrid nanofluids with those of all the corresponding mono nanofluids in identical conditions are required to establish whether the hybrids show values proportional to the amount of each single nanoparticle or if the combination of nanoadditives causes the appearance of synergistic effects.

The B_4C and TiB_2 mono nanoadditives and the $TiB_2:B_4C$ 20:80 wt% hybrid nanoadditive are used for the first time to design the corresponding 2 wt% nanofluids based on a propylene glycol:water 20:80 wt% mixture. SEM analyses of the nanopowders exhibit quasi-spherical B_4C nanoparticles with radii close to 60 nm and sheet-like TiB_2 nanoparticles with lengths/widths generally less than 1 μm .

The TiB_2 mono nanofluid presents the highest density increases, 1.5%, and the highest isobaric heat capacity decrease, 1.7%, with respect to the base fluid. The B_4C mono nanofluid shows the highest thermal conductivity enhancement, 6.0%. The density, isobaric heat capacity and thermal conductivity of the $TiB_2:B_4C$ hybrid nanofluid present intermediate values between those of the mono nanofluids. The experimental values of these properties were compared with those modelled by means different classical models showing good agreements.

All nanofluids evidence a shear thinning behaviour with a second Newtonian plateau at the high analysed shear rates. The B_4C mono nanofluid shows the highest dynamic viscosity increases with respect to the base fluid, 51–54% while the $TiB_2:B_4C$ hybrid nanofluid show non intermediate variation in viscosity in relation to that for the explored mono nanofluids, and present the lowest increase, 10–14%. Possible interactions between spherical and sheet-like nanoparticles could explain this lower resistance to flow. The VFT equation correlates well the dependence of dynamic viscosity on temperature.

The surface tension of the tested mono and hybrid nanofluids does not differ much from the pure base fluid whereas the electrical conductivity of the B_4C mono nanofluid shows much higher enhancements

over the base fluid than that of the TiB_2 mono nanofluid. The $TiB_2:B_4C$ hybrid nanofluid shows similar electrical conductivity behaviour than TiB_2 mono nanofluid, which suggests a negative effect of the larger TiB_2 particles.

The reported physical profile allows to conclude that the $TiB_2:B_4C$ hybrid nanofluid presents good perspectives as a heat transfer fluid, showing thermal conductivities close to those of the most conductive nanofluid (the B_4C mono nanofluid) and the lowest viscosity increases (and therefore pumping power). For electrical applications, the B_4C mono nanofluid brings huge electrical conductivity improvements.

Declaration of Competing Interest

There is no conflict of interest.

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